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Comparison of strontium isotope ratios in Mexican human hair and tap water as provenance indicators

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ABSTRACT

Deceased undocumented border crossers are some of the most difficult individuals to identify due to the inability to narrow down the region of origin and therefore to obtain family reference samples for DNA comparison. The isotopic compositions of various body tissues have been demonstrated to be useful biomarkers for tracking locations and movements to aid in the identification of human remains. This study closes the large spatial gap of available $^{87}Sr/^{86}Sr$ ratios from North America in tap water and presents the first $^{87}Sr/^{86}Sr$ human tissue-based ratios from Mexico. The 101 hair samples from 32 locations in Mexico range in $^{87}Sr/^{86}Sr$ ratios from 0.70424 to 0.71613 ($\Delta Sr_{max-min}$ = 0.01189). Furthermore, 151 tap water samples from 51 locations range between 0.70404 to 0.71385 ($\Delta Sr_{max-min}$ = 0.00981). Overall, small variations in the hair and tap water samples collected from individual locations were recorded ($\Delta Sr_{max-min}$ = 0.00041 and 0.00034 respectively). Despite the fact that Mexico is one of the largest bottled water consumers in the world, the $^{87}Sr/^{86}Sr$ ratios of human hair and tap water correlated strongly (R^2 = 0.87 for location averages and R^2 = 0.80 when using individual data points). These data represent a valuable resource for identifying the provenance of human remains.

1. Introduction

Migration and refugee movements have become major humanitarian crises around the world. Over 7000 individuals have perished along the Mexico - United States border since 1998 and the numbers continue to rise [1,2]. Many of the undocumented border crosser remains found, especially those recovered in Arizona, originated from Mexico [3–5]. Undocumented border crossers are difficult to identify and many are unlikely to ever be identified. One of the issues associated with the lack of identification is the inability to narrow down the region of origin and therefore identify family members in order to obtain reference samples for DNA comparison.

Because human tissues reflect the intake of biologically available strontium (Sr) during the time of formation, strontium isotopes are especially useful for studying migration patterns and predicting probable regions of origin of unidentified individuals [6,7]. Strontium isotopes have been used for studies in archeology, past conflicts and animal migration in various regions of the world, including Africa [8,9], Asia [10], the Americas [11–15], and Europe [16–18]. More recently, forensic anthropologists have discovered the usefulness of strontium isotope ratios for provenancing unidentified individuals from various contexts, including forensic casework and undocumented border crossers found along the Mexico - United States border [19–21]. Isotope analysis cannot be used to predict an individual's exact location of origin, but can add critical information to the identification process. Isotope analysis to narrow down geographic origins of unknown individuals is most useful when combining several isotope systems, such as the use of oxygen and strontium in combination [21].

To date, limited isotopic research has been conducted on bioavailable elements in modern Mexican environments for application to unidentified human remains. Large scale efforts thus far only focused on the oxygen, hydrogen, and deuterium excess in ground and tap water [22,23]. Some spatially limited archeological studies have explored lead and strontium isotopes for monitoring ancient human migration [24,25]. The limited available data is a major concern when trying to answer research

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questions from various fields, such as forensic anthropology, archeology, hydrology and food forensics. As an initial contribution to addressing this deficiency, this study reports $^{87}Sr/^{86}Sr$ ratio in the environment as determined in human hair and tap water from Mexico.

As Mexico is known to have one of the highest bottled water consumption rates in the world [26,27], this study will also aim to examine the impact on $^{87}Sr/^{86}Sr$ ratios of high tap water intake into the human body. Bottled water in the United States is often of local origin and hence has oxygen (O) and hydrogen (H) isotope signatures very similar to the local waters [28]. A similar relationship is expected for Mexican bottled waters but is not yet validated. This work assesses if tap water is a good proxy for the local bioavailable Sr and hence mimics the $^{87}Sr/^{86}Sr$ ratio found in the human body, in this case hair.

Due to the fact that the modern human's diet is becoming increasingly globalized, researchers have argued that isotopic signatures are becoming more and more homogenized [29,30] and may therefore inhibit scientists from distinguishing populations. This was most recently been demonstrated in the Netherlands, where there is no correlation between $^{87}Sr/^{86}Sr$ ratios in human enamel samples and tap water and that $^{87}Sr/^{86}Sr$ of water could not be used as a proxy in a mobility contexts [31]. For Mexico, over half of the food product imports originate from the United States [32]. Based on the fact that food intake is a major contributor to $^{87}Sr/^{86}Sr$ ratios found in human hair, this study will examine whether or not the "supermarket" diet significantly influences Mexican human hair ratios.

Researchers have argued that the $^{87}Sr/^{86}Sr$ ratios in human hair may be skewed due to exogenous signatures embedded in human hair, such as bathing water and dust [33–36]. Any "contamination" is in fact local and therefore will not significantly influence the goal of characterizing the bioavailable Sr of a region. It is important to note, however, that human hair from forensic cases are most often contaminated by the burial contexts and are not suitable to be used for region of origin predictions. This phenomenon has also been observed in cases of undocumented border crossers found in the Arizona desert [23].

This study presents and contrasts the $^{87}Sr/^{86}Sr$ ratios found in human hair and tap water from Mexico and compares the data to the United States. The objective is to provide the first large scale tap water and human tissue-based Sr isotope reference dataset for Mexico as a complementary dataset to the rest of North America.

1.1. Strontium isotopes ($^{87}Sr/^{86}Sr$)

Strontium has four naturally occurring stable isotopes with ^{87}Sr being the decay product of ^{87}Rb (rubidium, [37]). The isotopic composition of Sr is reported as the $^{87}Sr/^{86}Sr$ ratio because of their similar relative abundances. Strontium isotopes in the environment largely represent the underlying bedrock and predominantly vary based on the age of the rock and the initial amount of rubidium present, e.g., higher Sr isotope ratios are associated with older and Rb-rich rocks/sediments [38]. Strontium is removed from the bedrock through (differential) weathering and is transferred to our food chain though soil, flora and fauna. Strontium substitutes in biological systems for calcium and retains local geological information from the primary consumer to high-level consumers (such as humans) [39]. Not only is mass dependent fractionation negligible in strontium isotopic analyses due to its high atomic mass, it is also corrected for through the routine normalization procedures during the mass spectrometry measurements [38]. Therefore, in contrast to light isotopic systems such as oxygen and hydrogen, measured $^{87}Sr/^{86}Sr$ record no isotopic fractionation due to biological processes. Furthermore, as explained in detail by Bataille et al. [40], the regional Sr isotope signatures are predictable because they are controlled by geological variations, i.e. age and rock composition. The isotopic signature is passed from the underlying bedrock to the soil and consequently the bioavailable Sr is taken up by the food chain. The $^{87}Sr/^{86}Sr$ ratio of tissues reflect the intake of bioavailable $^{87}Sr/^{86}Sr$ ratios during the time of tissue formation so that strontium isotopes are commonly used for migration studies and region of origin predictions [7,41,42].

1.2. Keratin as a record of bioavailable Sr

The human head typically has between 80,000–150,000 hair follicles. These small pockets in the skin and the blood vessels around them create more cells, which then grow hair [43]. Once the hair exits the scalp, it dies off and elemental exchange between hair and blood stops. The biogenetic dietary information initially inherited from the blood is retained. While the organ undergoes a life-long cycle of growth, regression and a resting period before it falls out, it has been determined that, on average, human hair grows $\sim$1 cm per month. Therefore, hair retains a longitudinal record of isotope signatures and is thought to be useful to examine the recent geographic movement of an individual prior to their death [36,44,43]. Keratin tissues, such as hair, are, however, known to potentially incorporate environmental contamination, such as bathing water or dust [44,45]. For this study, however, even if there is significant environmental contamination it would be of local origin. Nonetheless, it is important to note that it is always preferable to compare the same body tissues (e.g. develop a baseline database of Mexican human teeth to compare with teeth of undocumented border crossers). However, due to issues of practicality and the time constraints, hair was chosen as the human proxy.

1.3. Water as a record of bioavailable Sr

Like many nations around the globe, Mexico faces a constant challenge to provide sufficient drinkable tap water resources to its citizens. The Mexican authorities and other institutions like the Inter-American Development Bank have attempted to resolve the issue of contamination in the Mexican river and lake system and improve the sanitation system [27,46,70]. However, there is still a lack of clean tap water throughout the country. Over 30 million Mexican citizens lack high quality water services or have limited access to water and Mexico is one of the largest bottled water consumers in the world [27,26,47,70]. Strontium isotope ratios of water have commonly been used to examine ecosystem processes and answer various hydrological and geological questions, such as surface and ground water interactions, source mixing, contamination, provenancing of water, and mineral weathering [48–50,51,52]. It is commonly assumed that the $^{87}Sr/^{86}Sr$ signature is directly reflective of the Sr found in the local ecosystem and local bedrock. However, this is not the case for water as water will mimic the $^{87}Sr/^{86}Sr$ of bedrock that is more soluble through weathering (such as limestone) [53,54]. Therefore, bedrock more susceptible to weathering will have a greater influence on the $^{87}Sr/^{86}Sr$ ratios of water than less soluble bedrocks [55]. Overall, ground water and therefore tap water is expected to reflect a wide range of Sr isotope ratios as it will reflect a mixture of recent rainwater and weathered bedrock [31]. This study is designed to assess how well drinking water reflects the bioavailable Sr taken up by hair of local individuals.

2. Methods

2.1. Sample acquisition and preparation

During a six-week period between June and July 2018, a total of 101 discarded human hair and 151 tap water samples were collected throughout Mexico by the lead author. The chosen sample locations were selected to cover the geographic variation across Mexico. The population sizes forthe sampled hair locations ranged from approximately 1500 to 200,000 inhabitants. For tap water, the population sizes ranged from approximately 1500 to 21 million. The hair collection occurred in 32 locations across 22 states and was completely anonymous (red and green dots in Fig. 1). Hair samples, but no water samples, were collected from Puerto Morelos, Quintana Roo (n = 3) and Saltillo, Coahuila (n = 1) (green dots in Fig. 1). No information was collected regarding the sex, age, dietary preferences, or travel history. Therefore, this study works under the assumption that the hair samples belonged to local individuals. At least three samples per location were gathered except for La Paz, Baja California Sur and Saltillo, Coahuila, where only one sample was collected. Since the hair samples were collected from the floors of barber shops, only bunches of hair were collected to ensure that the samples were not cross-contaminated. Each individual hair sample was examined to ensure that texture and color was homogenous. The samples were stored in separate paper envelopes in airtight zip-log bags at room temperature. The cleaning procedure adopted to remove dust and grease from the human hair samples is outlined in the Supporting Information 1.

Tap water samples were obtained from 30 of the 32 hair sample locations (for the remainder of the study named “hair locations”). Tap water samples were collected from 21 further locations and 5 additional states (blue dots in Fig. 1). Samples were collected from local tap water sources by running cold water for approximately 10 s before filling a clean 30 mL low-density polyethylene (LDPE) vial [56]. The cleaning procedure for the vials can be found in the Supporting Information 2.

2.2. Sample analysis

All samples were analyzed for their strontium isotope composition at the Vrije Universiteit Amsterdam, using a ThermoFinnigan Triton Plus thermal ionization mass spectrometer (TIMS). The protocols for Sr extraction and loading can be found in the Supporting Information 3. Procedural blanks were prepared with the human hair and tap water samples and analyzed alongside reference material. The reference material included the Standard Reference Material® 987 and an internal tooth standard from the Vrije Universiteit Amsterdam. Isotope ratios were corrected for mass-fractionation to $^{86}Sr/^{88}Sr = 0.1194$. The NBS987 standard gave an $^{87}Sr/^{86}Sr$ mean of 0.710256 ± 0.000009 (n = 45). The procedural blanks contained on average 37 pg strontium (n = 26).

2.3. Statistical and geospatial methods

Spatial representation of the data was carried out using ArcGIS 10.6 [57]. The geostatistical analyses for this study were performed using the ArcGIS Geostatistical Analyst toolbox and Geostatistical Wizard [57]. The tool “Global Moran’s I Spatial Autocorrelation” was used to determine the spatial autocorrelation based on the locations of the data points (hair and tap water sampling locations) and their corresponding isotopic ratios. The weight matrix was determined using inverse distance. The resultant Moran’s Index provided information as to whether the isotopic data is random, dispersed, or clustered in a spatial context [57]. Normality was tested using the D'Agostino-Pearson omnibus normality test. P-values were considered significant at the $\alpha = 0.05$ level. The nonparametric Mann-Whitney *U* test was employed to compare the different datasets.

3. Results

A total of 101 hair and 151 tap water samples were analyzed for their $^{87}Sr/^{86}Sr$ ratios. The basic descriptive statistics can be found in Table 1 and the results for all $^{87}Sr/^{86}Sr$ ratios of human hair and tap water measurements in Supporting Information 4 and 5, respectively. Sr isotope ratios for the human hair samples ranged from 0.70424 to 0.71613, with a range ($\Delta\ ^{87}Sr/^{86}Sr$) of 0.01189. The tap water samples varied from 0.70404 to 0.71385 ($\Delta\ ^{87}Sr/^{86}Sr = 0.00981$). The mean ratio for the hair samples was higher (0.70706) than for the tap water samples (0.70665 for all 51 locations and 0.70663 for the 30 hair locations) but all averages are

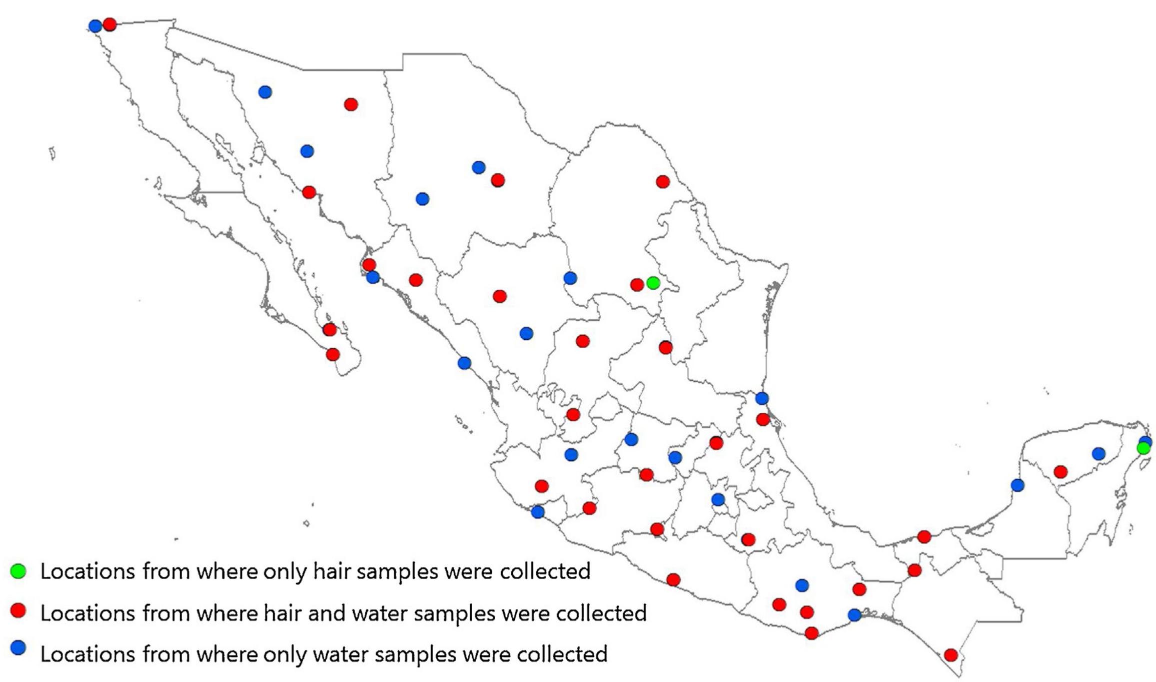


Fig. 1. Map of Mexico showing the sampling locations (green: n = 2, red: n = 30, blue: n = 21). Color figure available online (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

Table 1
Basic descriptive statistics for human hair and tap water samples.

	Human hair-all data	Human hair-location averages	Tap water-all data	Tap water-location averages	Tap water from hair locations-all data	Tap water from hair locations-location averages
N	101	30	151	51	88	30
Min	0.70424	0.70429	0.70404	0.70408	0.70404	0.70408
Max	0.71613	0.71239	0.71385	0.71347	0.71385	0.71347
Mean*	0.70706	0.70698	0.70665	0.70670	0.70663	0.70673
SD	0.00180	0.00178	0.00185	0.00183	0.00205	0.00212
Range	0.01189	0.00810	0.00981	0.00939	0.00981	0.00939

* all are within error.

within 1 SD (0.0018). Averaging the ratios recorded for each sampling location, calculating a so-called location average, reduces the range of the hair ratios 0.70429 to 0.71239, with an average of 0.70698. The tap water average location minimum and maximum ratios were the same for the 51 locations as for the 30 hair locations, ranging from 0.70408 to 0.71347. The mean $^{87}Sr/^{86}Sr$ of tap water from the hair locations (0.70673) is within error of the mean $^{87}Sr/^{86}Sr$ of tap water from all locations (0.70670).

The distribution of $^{87}Sr/^{86}Sr$ ratios is reported in Fig. 2. The $^{87}Sr/^{86}Sr$ ratios for both human hair and tap water samples are not normally distributed. Normality was tested using the D'Agostino-Pearson omnibus normality test (human hair: $K^2 = 52.5$, p-value < 0.001; tap water: $K^2 = 44.0$, p-value < 0.001).

A visualization of the spatial distribution of $^{87}Sr/^{86}Sr$ ratios of the hair and tap water samples can be found in Fig. 3. The Moran's Index for the hair samples yielded 0.344, with a p-value of 0.186. Given the z-score of 1.323, the pattern does not appear to be significantly different from random. In contrast, the tap water samples yielded an index of 0.768 with a p-value of 0.000. Given the z-score of 6.467, there is a less than 1% likelihood that this clustered pattern could be a result of random chance.

For both hair and tap water samples, the lowest Sr isotope ratios appear along the west coast and Sierra Madre mountainous range, mainly in the states of Guanajuato, Jalisco and Michoacán. The highest $^{87}Sr/^{86}Sr$ ratios were found in the states of Oaxaca and the north-west of Mexico, specifically Tijuana (Baja California) (Fig. 3a and b). The lowest $^{87}Sr/^{86}Sr$ ratios recorded in human hair was documented in Moroleón, Guanajuato (0.70424), and the highest in Miahuatlán de Porfirio Díaz, Oaxaca (0.71613) (Table 1, Fig. 3a). The lowest $^{87}Sr/^{86}Sr$ ratios of tap water was found in El Grullo, Jalisco (0.70404), and the highest in Miahuatlán de Porfirio Díaz, Oaxaca (0.71385) (Table 1).

Standard deviations were calculated for each location. The results for the human hair samples, tap water samples, and the tap water samples from hair locations can be found in Table 2. A spatial visualization of the standard deviations for the human hair and tap

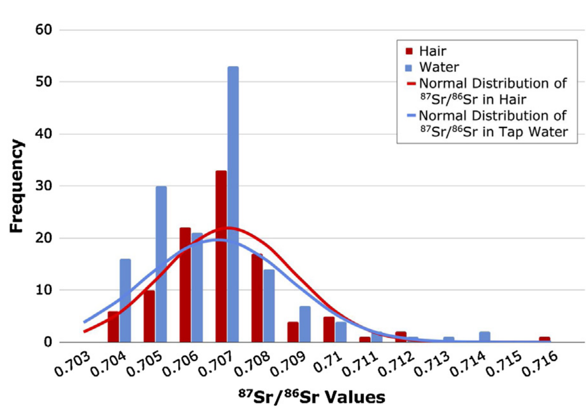


Fig. 2. Distribution of human hair and tap water $^{87}Sr/^{86}Sr$ ratios in Mexico. Color figure available online.

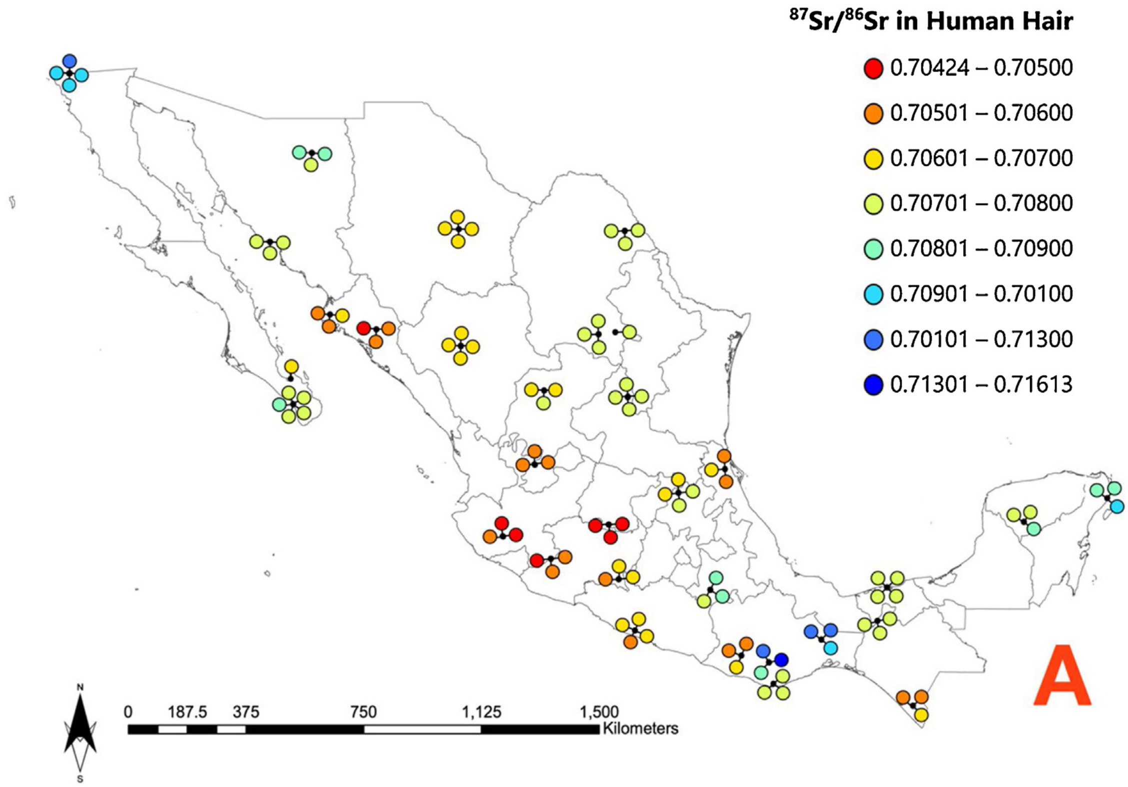
water sample locations can be found in Fig. 4. The smallest standard deviation (0.000009) in the human hair samples was recorded in Villa Unión, Coahuila (n = 3) and the highest in Miahuatlán de Porfirio Díaz, Oaxaca (0.00372, n = 3). When removing the latter location from the dataset, the maximum standard deviation drops to 0.00130 (Paraíso, Tabasco, n = 4). On average, the locations showed a standard deviation of 0.00041 (0.00029 when excluding Miahuatlán de Porfirio Díaz).

Tap water samples were analytically indistinguishable at Tampico, Tamaulipas (n = 3). The highest standard deviation was 0.00335 in Tijuana, Baja California (n = 3). Excluding Tijuana, the standard deviation maximum was 0.00286, recorded in Miahuatlán de Porfirio Díaz, Oaxaca (n = 3). The average standard deviation for tap water samples was 0.00034, 0.00028 when removing Tijuana from the dataset. The highest standard deviation at the hair sample locations was at Miahuatlán de Porfirio Díaz, Oaxaca. The average standard deviation was 0.00027. A justification for removing the two abovementioned locations (Tijuana and Miahuatlán) will be presented in the discussion. Furthermore, as can be seen in Figs. 3, 4 and 6, both locations are clear outliers for the hair and the tap water samples, respectively.

4. Discussion

The $^{87}Sr/^{86}Sr$ ratios in Mexican human hair and tap water have a relatively large absolute range ($\Delta Sr_{max-min} = 0.01189$ and $\Delta Sr_{max-min} = 0.00981$, respectively) while, at the same time, showing limited variation within the samples collected from one location, typically $\Delta^{87}Sr/^{86}Sr$ = <0.0003. The Mann-Whitney *U* test showed that there was a significant difference (W = 8867, p-value = 0.029 for all data, W = 5004, p-value = 0.024 for the 30 locations with combined tap water – hair data) between the $^{87}Sr/^{86}Sr$ ratios of human hair and the $^{87}Sr/^{86}Sr$ ratios of tap water. However, when using the location averages for the 30 hair locations, the Mann-Whitney *U* test showed no significant difference between the two datasets (W = 478, p-value = 0.378). The $^{87}Sr/^{86}Sr$ ratios in human hair and tap water correlated strongly to one another (Fig. 5a and b), but did not yield a 1:1 relationship. The correlation between the $^{87}Sr/^{86}Sr$ ratios of tap water and human hair yields an R^2 of 0.80. When using the averages calculated for each sampled location the correlation increases R^2 to 0.87.

Three main sources are considered to contribute to the $^{87}Sr/^{86}Sr$ ratios in human hair: exogenous, drinking water, and diet. Despite the fact that several studies have reported potential exogenous signatures embedded in human hair, most likely influenced by bathing in local tap water and from local dust [33–36], it is argued in this study, that the "contamination" would be of local origin and therefore not significantly offset the results in terms of potential provenance information. As previously mentioned, Mexico is known to be one of the highest bottled water consumers in the world. In many industrialized countries, such as the United States, bottled water is regionally sourced and therefore almost indistinguishable from the local tap water [58]. It is unknown if this is also the case for Mexico but from anecdotal evidence that the bottled



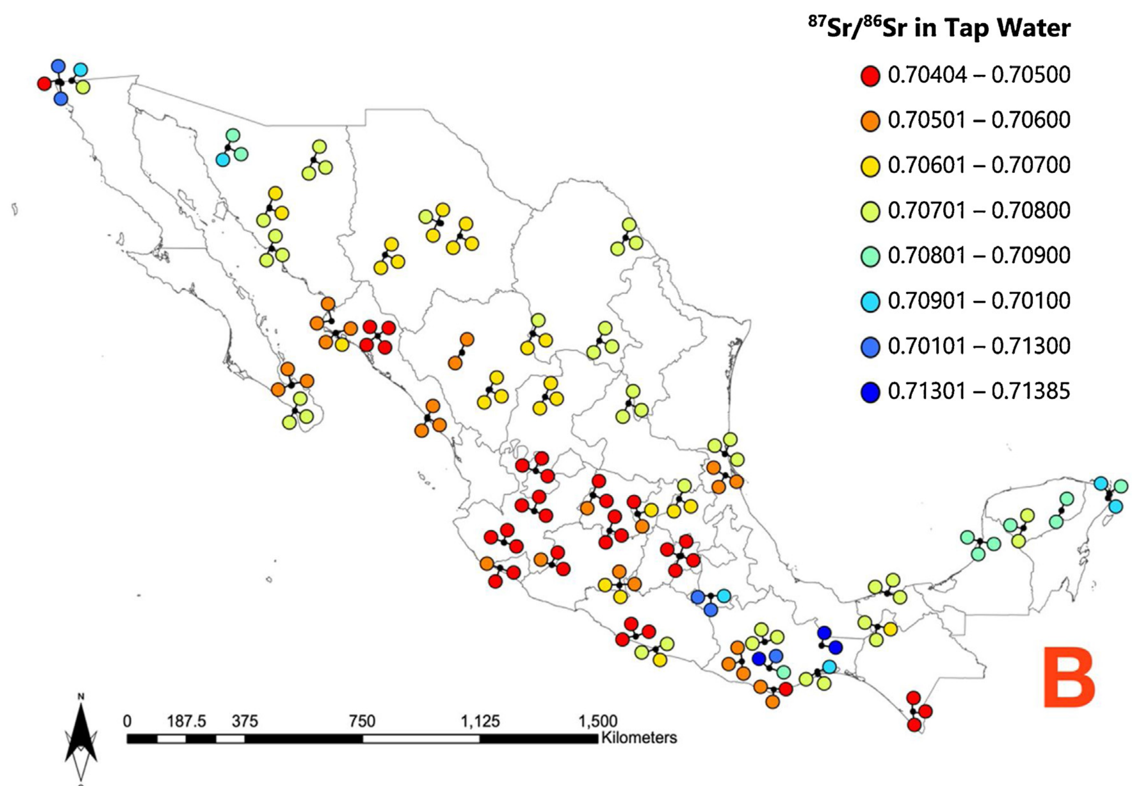


Fig. 3. Graphical distribution of modern human hair (A) and tap water (B) $^{87}Sr/^{86}Sr$ ratios in Mexico. Color figure available online.

Table 2
Standard deviation values for human hair and tap water.

	Human Hair	Tap Water from Hair Locations	Tap Water
Min	0.000009	0.000005	0.000002
Max	0.00372 / 0.00130*	0.00286	0.00335 / 0.00286#
Mean	0.00041 / 0.00029*	0.00027	0.00034 / 0.00028#

* These values exclude the location (Miahuatlán de Porfirio Díaz, Oaxaca) with the highest standard deviation within the collected samples.
# These values exclude the location (Tijuana, Baja California) with the highest standard deviation within the collected samples.

water used for drinking and cooking is mostly produced by large regional companies and transported to local communities. Future study of Mexican bottled water could potentially indicate the proportion of Sr in human body tissues derived from bottled and tap water. Even though the low Sr content in drinking water means that the bulk of Sr ingested by humans is from food [31], it is a significant finding of this work that the $^{87}Sr/^{86}Sr$ ratios of human hair and tap water are strongly correlated.

While more rural communities grow most of their own produce there will, nonetheless, be some influence from the so called "supermarket diet". Certain foods will be transported to the communities and therefore could, potentially, lead to more homogenous isotopic signatures recorded in human tissue. This effect has been demonstrated most recently by Kootker et al. [31] examining the relationship of $^{87}Sr/^{86}Sr$ ratios in Dutch tap water and human tooth samples. While a direct comparison with the Netherlands is invalid due to the significant socio-economic differences between the countries, this study clearly establishes that regional geology has a major influence and hence it can be concluded that the supermarket diet does not significantly skew the data in Mexico.

Overall, the human hair samples are more radiogenic than the tap water counterparts in the same locations up to Sr isotope ratios of circa 0.708. Above 0.708, the Sr isotope composition of tap water is more radiogenic than that of the hair samples. This phenomenon could be explained by ambient dust contamination of hair coupled with the fact that low ^{87}Sr originates from volcanic rocks that contain Sr-rich plagioclase with low Rb and hence low $^{87}Sr/^{86}Sr$. Breakdown of plagioclase will release Sr with lower $^{87}Sr/^{86}Sr$ than the bulk rock that will more readily enter the ground water, leaving soil with higher $^{87}Sr/^{86}Sr$ (Bain & Bacon 1994; Jung et al. 2004). In contrast, silica rich crustal rocks preferentially break down potassium feldspar and micas with higher $^{87}Sr/^{86}Sr$ into the water leaving soils with lower $^{87}Sr/^{86}Sr$. The presence of Cretaceous carbonates in parts of Mexico is another contributing factor. Sr-rich marine carbonates have $^{87}Sr/^{86}Sr$ derived from seawater and have fluctuated between 0.707 and 0.709 over the last 500 Myr, with Cretaceous rocks close to 0.708. Redistribution of marine derived Sr in rainwater and as dust from the carbonate rocks will drive bioavailable Sr towards $^{87}Sr/^{86}Sr$ ratios of 0.708 to 0.709. A more systematic evaluation of the causes of the isotopic offset between water and human tissues is warranted, taking the role of the local climate and hence weathering processes into account. Further research is also required on the uptake of Sr into other human tissues, such as bones and tooth enamel, in order to evaluate the predictive value of human hair as well as tap water into different tissue types.

4.1. Geospatial patterns

The Moran's Index suggests hair samples were randomly spatially dispersed while the tap water samples appear to be clustered. There are two main reasons why the results of the Moran's Index may be contradictory. Prior to the sample collecting trip, a geological map of Mexico was used to choose sampling locations. A total of 30 locations were chosen by geological variation in order to obtain a relatively complete coverage of Mexico. These were the locations from where hair samples were collected. It is unexpected that the Moran's Index recognizes spatial patterns from merely 30 locations that are primarily based on geology (and are therefore not necessarily spatially clustered). Since the samples were collected on a road trip, more tap water samples were collected from locations that lay geographically between the 30 hair locations. Therefore, more spatial clustering can be expected since more locations were sampled, which are also in closer proximity to one another. The spatial patterns and the relationships of the $^{87}Sr/^{86}Sr$ ratios to the underlying bedrock will be examined in the future. Overall, the standard deviations of both human hair and tap water were relatively small for individual locations, giving confidence in the sampling strategy and emphasizing that these locations are supplied with water from similar sources. Specifically, the $\Delta^{87}Sr/^{86}Sr_{total}$ is 0.0018 for the datasets (Table 1) while the average $\Delta^{87}Sr/^{86}Sr_{locations}$ is <0.0003.

4.2. Variance in $^{87}Sr/^{86}Sr$

There was no correlation between the size of the local human population and the recorded SD in the Sr isotope data (for human hair, $R^2 = 0.0006$, or tap water, $R^2 = 0.0002$). The population sizes for the sampled hair locations ranged from approximately 1500 to 200,000 inhabitants [59,60]. For tap water, the population sizes ranged from approximately 1500 to 21 million [59,21]. The fact that population size did not influence the isotopic variation shows that the sampling strategy was successful, especially as it was designed to aid in predicting the region of origin of undocumented border crossers, who have traditionally originated from smaller and more rural locations. Nonetheless, it would be advisable to further investigate larger municipalities as a recent shift has been documented towards migration from more urban areas [61]. Furthermore, analyzing megacities such as Mexico City would provide more comprehensive information on the isotopic ranges in human hair within the entire Mexican population.

Miahuatlán de Porfirio Díaz (Oaxaca) recorded the highest $^{87}Sr/^{86}Sr$ ratios and variance for human hair and the second highest in tap water. The highest SD value in the tap water data was documented in Tijuana (Baja California), which also showed the highest SD values for oxygen and hydrogen in a previous study examining the same tap water samples [23]. This variation is probably due to the city's reliance on multiple water sources (e.g. the Colorado River) [62,63]. Despite the limited number of samples per location (n = 2–5) the Sr isotope variance in water and hair samples show a general correlation (Fig. 4a and b), with an R^2 of 0.52 when the location of Tecate (Baja California) was disregarded.

4.3. Comparison with the United States

This research aims to provide a comparative baseline for provenancing human remains found along the Mexico-United States border. It is therefore pertinent to compare the Mexican data to data reported from the United States [33,55]. By comparing data, it will potentially be possible to determine whether an individual

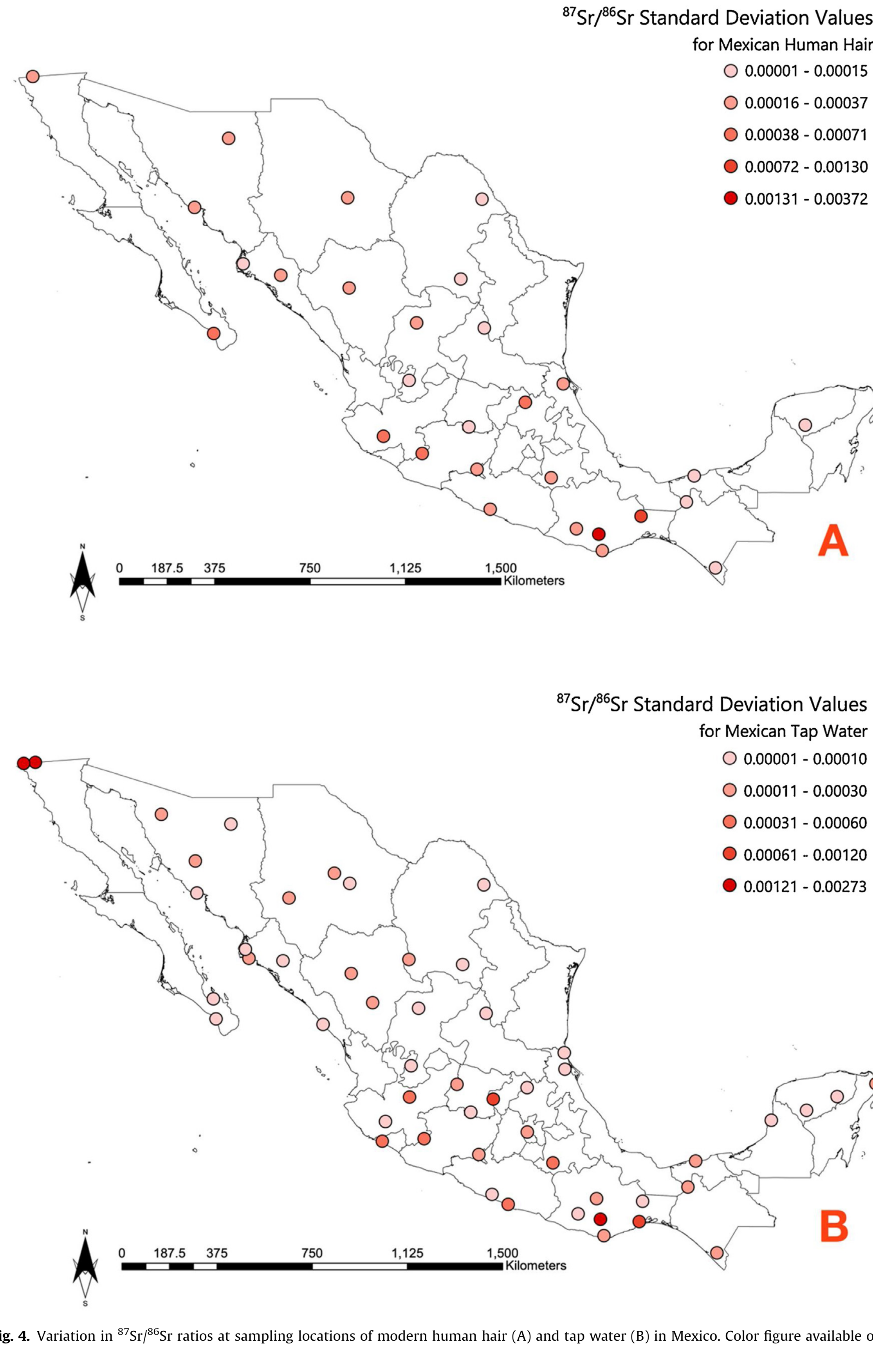


Fig. 4. Variation in $^{87}Sr/^{86}Sr$ ratios at sampling locations of modern human hair (A) and tap water (B) in Mexico. Color figure available online.

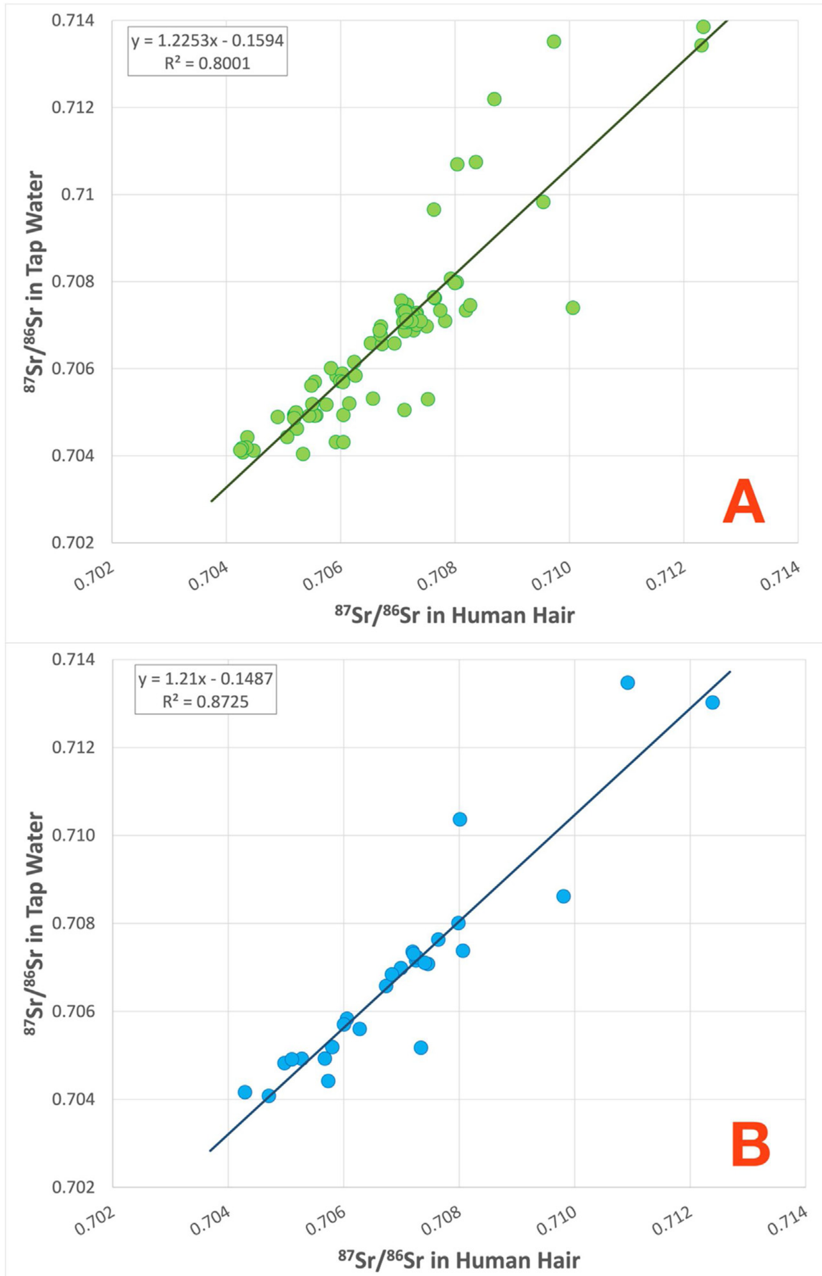


Fig. 5. Relationship of $^{87}Sr/^{86}Sr$ ratios in human hair and tap water collected from the sample locations (n = 30).
A: all data ($^{87}Sr/^{86}Sr$ Tap Water = 1.23 * $^{87}Sr/^{86}Sr$ Human Hair -0.16; R^2 = 0.80)
B: location averages ($^{87}Sr/^{86}Sr$ Tap Water = 1.21 * $^{87}Sr/^{86}Sr$ Human Hair -0.15; R^2 = 0.87).

originated from Mexico or the United States or may have moved between the two countries.

Mann-Whitney *U* tests showed that there is a significant difference between the available hair and tap water data from the United States and the hair data and tap water data from Mexico (W = 9185, $p < 0.001$ for hair; W = 12534, $p < 0.001$ for tap water). Furthermore, the Mexican Sr isotope ratios for human hair and tap water, are, on average, lower compared to the United States (Fig. 6). For the hair samples, approximately 75% percent of the data fall below the lowest recorded ratios in the United States human hair dataset ([33], Fig. 6). Therefore, based on the current dataset, $^{87}Sr/^{86}Sr$ ratios below 0.707 are indicative of individuals of Mexican origin. Ratios above cannot be distinguished. In contrast, there is a complete overlap of the Mexican tap water and the United States tap water. The lowest ratios recorded for tap water in Mexico are comparable to the lowest ratios found in the United States (Fig. 6). Furthermore, the fourth quantile of the Mexican data reaches the median of the United States ratios.

This observation can be explained by the younger underlying bedrock in Mexico [64]. As explained in the introduction, lower $^{87}Sr/^{86}Sr$ ratios are associated with younger volcanic bedrock [37,39,50,65]. While these isotopic differences are certainly meaningful, it is important to note that the data from the United States include few samples from either of the coasts, which may somewhat bias the data. Based on the absence of sample locations in the western United States, where lower Sr isotope ratios are predicted in underlying bedrock [65,66], the observation of an isotopic difference in hair samples could be subject to change in the future. This illustrates the importance of using a multi-isotope

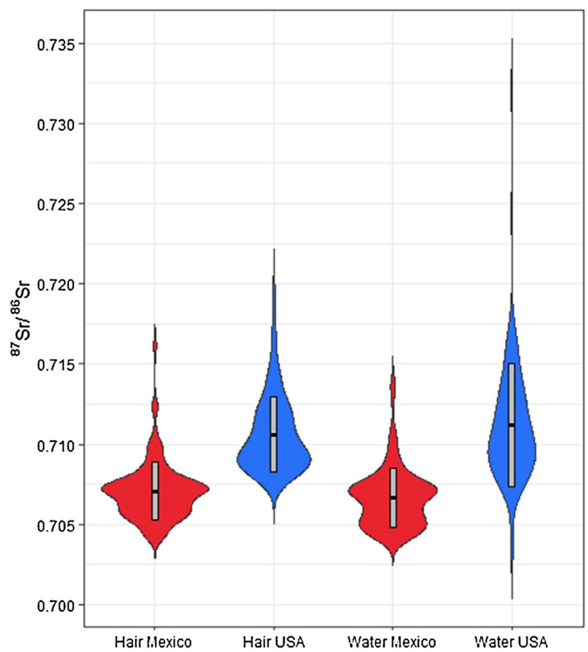


Fig. 6. Violin plot showing $^{87}Sr/^{86}Sr$ ratios in human hair and tap water from Mexico and the United States [33,55]. The mean ± standard deviation was added as a crossbar.

approach when provenancing (e.g. combine Sr and O isotopes). Nonetheless, considering that undocumented border crossers are usually found in contexts inconsistent with United States citizens, this research will be able to serve as a basis for provenancing unidentified individuals.

4.4. Additional isotope datasets

Limited isotopic research has been conducted into bioavailable elements in modern Mexican environments, with large scale studies limited to oxygen, hydrogen, and deuterium excess. There have been, however, some spatially limited Sr isotope studies. The $^{87}Sr/^{86}Sr$ ratios previously recorded for modern Mexican teeth range from ~0.7053 to 0.7080, falling within the ranges recorded in this study [67,68]. Hodell and colleagues [25] presented data on water, bedrock, soils, and plants for the Yucatan Peninsula, with $^{87}Sr/^{86}Sr$ ratios (0.7071 to 0.7099) covering the same range as reported here for human hair and tap water in the region (Fig. 3). Archeological human bone and tooth enamel from southern Mexico showed that the $^{87}Sr/^{86}Sr$ range of the samples (approximately 0.704 to 0.708) is the same as this study [69]. Overall, the limited available Sr isotope data from modern and archeological human tissues as well as environmental samples validate the spatial variation recorded in this study.

5. Conclusion

This study presents the first large scale modern human tissue based $^{87}Sr/^{86}Sr$ ratios from Mexico and has closed a large spatial gap in $^{87}Sr/^{86}Sr$ tap water ratios available in North America. The hair samples from 32 locations in Mexico define a range in $^{87}Sr/^{86}Sr$ ratios from 0.70424 to 0.71613 ($\Delta Sr_{max-min}$ = 0.01189). The tap water samples from 51 locations ranged from 0.70404 to 0.71385 ($\Delta Sr_{max-min}$ = 0.00981). A strong correlation (R^2 = 0.80) is observed between $^{87}Sr/^{86}Sr$ of human hair and tap water data and location averages correlated more strongly (R^2 = 0.87).

This study has shown that the $^{87}Sr/^{86}Sr$ ratios in human hair and tap water from Mexico have great potential to be used for provenancing studies, such as determining potential regions of origin of undocumented border crossers found along the Mexico - United States border. Nonetheless, it is pertinent to expand the research to other known body tissue samples in order to establish potential relationships of human hair to body tissues more frequently consulted in forensic anthropology. Following the example of Kootker et al. [31], dental enamel samples could be collected in order to determine the predictive strength of human hair as well as examine the question whether tap water can serve as an indicator for tissues that are not potentially exposed to external contamination.

Since Mexico is one of the highest bottled water consumers per capita in the world, the examination of Mexican bottled water could potentially yield indicators as to what degree human body tissues are influenced by diet, bottled water, and tap water. Lastly, this research will serve as a basis for various machine learning regression approaches, which will include multivariate models, to determine regional Sr isoscape for Mexico [23]. Following the approach of Bataille et al. [40], the resulting isoscapes will take into consideration various geological covariates, such as the underlying bedrock and weathering regime in order to enable future provenancing studies.

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CRediT authorship contribution statement

Saskia T.M. Ammer: Conceptualization, Investigation, Methodology, Formal analysis, Writing - original draft, Visualization, Project administration, Funding acquisition, Validation. **Lisette M. Kootker:** Conceptualization, Investigation, Methodology, Writing - original draft, Writing - review & editing. **Eric J. Bartelink:** Supervision, Writing - review & editing, Methodology. **Bruce E. Anderson:** Supervision. **Eugénia Cunha:** Supervision. **Gareth R. Davies:** Supervision, Conceptualization, Methodology, Writing - review & editing.

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Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at https://doi.org/10.1016/j.forsciint.2020.110422.

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